FORM PTO-1390 (Modified) (REV 11-98) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER: 13929/122944

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR): 09/786097

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP99/06333	27 August 1999	28 August 1998

TITLE OF INVENTION

USE OF POLYESTER RESINS FOR THE PRODUCTION OF ARTICLES HAVING GOOD PROPERTIES AS BARRIERS TO WATER VAPOUR

APPLICANT(S) FOR DO/EO/US

Catia Bastioli, Marco Foa, Giandomenico Cella, Giovanni Floridi, Fernanda Farachi, and Tiziana Milizia

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
   a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210).
8. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
   a. ☒ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). (Unexecuted)
11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

Items 13 to 20 below concern document(s) or information included:

13. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☒ Certificate of Mailing by Express Mail
20. ☐ Other items or information:

1) Transmittal letter;
2) One checking totalling $990.00 to cover the filing fee for patent appln;
3) Notice Informing the Applicant of the Communication of the International Appln. to the Designated Offices (Form PCT/IB/308);
4) Information Concerning Elected Offices Notified Of Their Election (Form PCT/IB/332 (Sept. 1997)); and
5) Return postcard

U.S. APPLICATION NO (IF KNOWN, SEE 37 CFR	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/786097	PCT/EP99/06333	13929/122944

21. The following fees are submitted:.

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

		CALCULATIONS PTO USE ONLY
☐	Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO	**$1,000.00**
☒	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but Internation Search Report prepared by the EPO or JPO	**$860.00**
☐	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO	**$710.00**
☐	International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)	**$690.00**
☐	International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)	**$100.00**
	ENTER APPROPRIATE BASIC FEE AMOUNT =	**$860.00**
	Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☒ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).	**$130.00**

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	14 - 20 =	0	x $18.00	**$0.00**
Independent claims	1 - 3 =	0	x $80.00	**$0.00**
Multiple Dependent Claims **(check if applicable)**.			☐	**$0.00**
	TOTAL OF ABOVE CALCULATIONS =			**$990.00**
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) **(check if applicable)**.			☐	**$0.00**
	SUBTOTAL =			**$990.00**
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).			+	**$0.00**
	TOTAL NATIONAL FEE =			**$990.00**
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) **(check if applicable).**			☐	**$0.00**
	TOTAL FEES ENCLOSED =			**$990.00**
			Amount to be: refunded	$
			charged	$

☒ A check in the amount of **$990.00** to cover the above fees is enclosed.

☒ Please charge my Deposit Account No. **02-4467** in the amount of **$990.00** to cover the above fees.
A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **02-4467** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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Warren K MacRae
SIGNATURE

Warren K. MacRae
NAME

37,876
REGISTRATION NUMBER

02/28/01
DATE

Docket No.: 13929/122944

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re National Application of :)
Catia Bastioli, Marco Foa', Giandomenico Cella,
Giovanni Floridi, Fernanda Farachi, and Tiziana)
Milizia
International Appln. No. PCT/EP99/06333)
International Filing Dated: 27 August 1999

Filed Concurrently Herewith On February 28, 2001

For: **USE OF POLYESTER RESINS FOR THE PRODUCTION OF ARTICLES HAVING GOOD PROPERTIES AS BARRIERS TO WATER VAPOUR**

CERTIFICATE OF EXPRESS MAILING

"Express Mail" Mailing Label No.: EL715972925US
Date of Deposit: February 28, 2001

I hereby certify that the following:

- [x] This Certificate of Express Mailing
- [x] Transmittal Letter (Form PTO-1390) in duplicate (2 pp.)
- [x] Published International Application (23 pp.)
- [x] Unexecuted Declaration and Power of Attorney (Form PTO/SB/01) (5 pp.)
- [x] International Search Report (5 pp.)
- [x] Preliminary Amendment (2 pp.)
- [x] Check in the amount of $990.00 to cover filing fee
- [x] PCT Request (5 pp.)
- [x] PCT Demand (4 pp.)
- [x] Notification Concerning Submission of Transmittal of Priority Document (Form PCT/IB/304) (1 p.)
- [x] Notice Informing The Applicant Of The Communication Of The International Application To The Designated Offices (Form PCT/IB/308) (2 pp.)
- [x] Information Concerning Elected Offices Notified Of Their Election (1 p.)
- [x] International Preliminary Examination Report (Form PCT/IPEA/409) (8 pp.)
- [x] Information Disclosure Statement (2 pp.)
- [x] PTO-1449 Form to Information Disclosure Statement including references
- [x] Return postcard

are being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR Section 1.10 on the Date of Deposit indicated above in an envelope addressed to the Commissioner for Patents, Box PCT, Washington, D.C. 20231.

Gregory Small

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268993

Express Mail Label No. EL715972975US
Docket No.: 13929/122944

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re National Application of :
Catia Bastioli, Marco Foa' Giandomenico Cella, Giovanni Floridi, Fernanda Farachi, and Tiziana Milizia
International Appln. No. PCT/IT99/06333
International Filing Dated: 27 August 1999

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New York, New York
February 28, 2001

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

Sir:

Please amend the above-identified application as follows:

In The Claims

Please substitute the attached annexes to the International Preliminary Examination Report for pages 13-16 of the published PCT Application WO 00/12627.

Please amend claims 5, 6, 7, 8, 9, and 10 as follows:

Claim 5, line 1, change "any one of the preceding claims" to --claim 1--.

Claim 6, line 1, change "any one of the preceding claims" to --claim 1--.

Claim 7, line 1, change "any one of the preceding claims" to --claim 1--.

Claim 8, line 1, change "any one of the preceding claims" to --claim 1--.

Claim 9, line 1, change "any one of the preceding claims" to --claim 1--.

Claim 10, line 1, change "any one of the preceding claims" to --claim 1--.

Remarks

Claims 5, 6, 7, 8, 9, and 10 have been amended to eliminate multiple dependency and thereby minimize claim fees.

Respectfully submitted,

By: 

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Use of polyester resins for the production of articles having good properties as barriers to water vapour

The present invention relates to the use of biodegradable polyester resins in the production of formed articles having good properties as barriers to water vapour.

The water-vapour barrier properties of biodegradable polymers developed in recent years are quite poor.

For example, polyesters such as polyhydroxybutyrate-valerate, polylactic acid, polyglycolic acid, polycaprolactone, polybutylene succinate, copolymers such as polybutylene adipate-co-terephthalate, polyester-amides such as polybutylene adipate-co-caprolactam, polyvinyl alcohol, ethylene-vinyl alcohol copolymers, polyesters-urethanes, and esters of cellulose and regenerated cellulose have permeabilities to water vapour greater than 300 gx30μm/m^2 per day at 38°C and 90% relative humidity (RH) (Lyssy method).

The poor barrier properties can be related to the fact that these polymers have good biodegradability which, in order for the bacterial action to be performed advantageously, means that the polymer should be wettable and hence contains polar groups in its structure with a consequent reduction in its water-vapour barrier properties since the polar groups increase the solubility of water in the polymer and hence its permeability to water vapour.

High permeability to water vapour considerably limits the fields of use of biodegradable polymers such as the above-mentioned aliphatic polyesters or copolyesters, particularly where good biodegradability and low permeability to water would be very desirable.

Fields of use in which there is a particular need for biodegradable materials having good water-vapour barrier properties are, for example, the hygiene field (so-called non-breathable nappies, that is to say, nappies with a low transpiration value, similar to the nappies which are in use with a backsheet of polyethylene and non-woven polypropylene fabric), multi-layer and non-multi-layer food packaging based on laminated milk cartons, mulching of soils where the evaporation of water through materials is to be as limited as possible, containers for soil for growing plants in greenhouses, sacks for collecting grass cuttings which require reduced biodegradation rates by virtue of a lower wettability of the biodegradable film of which the sack is made, non-woven fabric which can provide a dry feel for nappies, fishing nets which must not undergo significant alterations due to water during the period of use, expanded products for packaging which requires moisture protection whilst remaining biodegradable, irrigation pipes for agriculture, products in contact with liquid foodstuffs, such as fast-food cups, plates and drinking straws, expanded trays for foodstuffs, blister packs for pharmaceutical products, nursery plant-pots through which moisture must not be able to pass and which must have a degradation process which does not interfere with the growth of the plants, hygiene products such as colostomy bags and the like, or blood containers, fibres for disposable products which can withstand water and a few washings, for disposable hosiery and garments, etc.

It has now been found, unexpectedly - in view of the outstanding permeability of aliphatic polyesters such as polybutylene adipate, polybutylene succinate, polyhexamethylene adipate and polybutylene adipate-co-terephthalate to water vapour - that the polyester resins defined below have good water-vapour barrier properties and,

at the same time, are sufficiently biodegradable in normal composting conditions and are therefore usable in applications in which such properties are required.

The polyester resins usable in the applications of the invention are formed by recurring units X = $[O-(CH_2)_n-OCO-(CH_2)_m-CO]$ and/or Y = $[O-(CH_2)_k-CO]$, where the half-sum of n + m is equal to or greater than 6 and k is a number equal to or greater than 6, or by copolymers comprising units and/or sequences having the formula $x_i[O-(CH_2)_{ni}-OCO-(CH_2)_{mi}CO]$; y_j $[O-(CH_2)_{kj}-CO]$ where:

i,j = 1-5; n_i = 2-22; m_i = 0-20; k_j = 1=21;

$\sum_{i=1}^{5} x_i + \sum_{j=1}^{5} y_j = 1$ and x_i and y_j vary between 0 and 1 and are molar fractions of the various units such that

$\sum_{i=1}^{5} x_i \left(\frac{n_i + m_i}{2}\right) + \sum_{j=1}^{5} y_j \; k_j \geq 6$, or by recurring units

Z= $[O-(CH_2)_a-OCO-(CH_2)_b-CO]$ where a=2-3 and b=7-11,

present in sufficient quantity to ensure good barrier properties and biodegradability of the resins in the production of products in which a permeability to water vapour of less than 350 gx30μm/m^2 per day at 38°C and 90% RH and biodegradability in composting or burial conditions are required.

The products which can be produced from the polyesters as defined above can ensure permeability to water vapour of less than 350, more particularly less than 300, gx30μm/m^2 per day at 38°C and 90% RH.

The biodegradability of the products during composting or burial is sufficient to bring about their decomposition within the required periods of time.

More particularly, in the case of the products produced from the preferred polyester resins, the biodegradability is less than 30% in one month and more than 60% in six months, in accordance with DIN 54900, part II, or decomposition on 30μm film of less than 10% in 14 days and more than 90% in 6 months, in accordance with the method described in "Journal of Environmental Polymer Degradation", Vol. 4, No. 1, 1996, p. 55-63, or in accordance with the burial test described in "Biodegradable Plastics, Practices and Test Methods" ASTM Subsection D-20.96.1 of Environmental Degradable Plastics, Version 4.0 Dec. 6 1990.

The polyester resins usable according to the invention have a mean numeral molecular weight greater than 10000 and a melting point (acceptable for industrial applications) of between 60 and 110°C.

Polyester resins with a mean numeral molecular weight of between 45000 and 70000 have been found particularly advantageous for use according to the invention.

There is not the slightest reference in the literature either to the barrier properties, particularly to water vapour, of the polyester resins falling within the general formula given above, or to their good biodegradability by decomposition.

The use of the above-mentioned polyester resins in applications which require a low permeability to water vapour (below the value indicated above) combined with a biodegradability during composting compatible with the standards in use is novel and constitutes the subject of the present invention.

Examples of applications in which the polyester resins according to the invention are particularly useful are:

- coatings produced by extrusion-coating with good water-barrier properties, particularly for the packaging of fresh milk and diary products, of meat, and of foods with high water content,
- multi-layer laminates with layers of paper, plastics material or paper/plastics material, aluminium and metallized films in general,
- films as such, and multi-layer films with other polymer materials,
- sacks for organic refuse and for grass cuttings with periods of use longer than 1 week,
- single-layer and multi-layer food packaging, particularly containers for milk, yoghurt, cheeses, meat and beverages, in which the layer in contact with the food or beverage is formed by the polyester,
- composites with gelatinized starch, destructured starch, native starch in the form of a filler, or complexed starch,
- mono-directional or bi-directional films,
- semi-expanded and expanded products produced by physical and/or chemical means, by extrusion, injection, or agglomeration of pre-expanded particles, from materials constituted by the polyester as such, from blends, or from filled materials,
- expanded sheet and expanded containers for foods, (fruit, vegetables, meat, cheeses) for drugs, and for fast-food,
- fibres, fabrics and non-woven fabrics in the hygiene, sanitary and clothing fields,
- outer non-woven fabric and/or film, front tapes for increasing the thickness of the backsheet in critical points, and adhesive strips, for the production of nappies,
- composites with mineral and vegetable fillers with various form ratios,

- extruded or thermoformed sheets and profiles in the field of food and fast-food packaging (drinking straws, cups, trays, etc.),
- bottles for the food, cosmetics and pharmaceutical fields,
- fishing nets,
- containers for fruit and vegetables,
- irrigation pipes in the agricultural field,
- products produced from blends with other biodegradable polymers (for example, polybutylene succinate, polycaprolactone, polyhydroxybutyrate-co-valerate, polyesters-amides, aliphatic-aromatic polyesters), for correcting the biodegradation rate, the processability, and/or the permeability to water of these latter polymers and the superficial properties such as migration phenomena of low molecular weight molecules,
- products produced from blends with non-biodegradable polymers.

Polyesters falling within the general formula given above can be produced by the polycondensation, in accordance with known methods, of a bicarboxylic aliphatic acid with 2-22 carbon atoms with a diol with 2-22 carbon atoms, selected in a manner such that the half-sum of the carbon atoms relating to the acid and to the diol is equal to or, preferably greater than 6, more preferably equal to 7, or by polycondensation of hydroxy-acids with 7-22, preferably 8-22 carbon atoms, or by ring-opening of the corresponding lactones or lactides; or by polycondensation of ethylen glycol with azelaic and sebacic acid.

Aliphatic-aromatic copolyesters, aliphatic-polyamide copolyesters, aliphatic-ether copolyesters, aliphatic-urea copolyesters or linear or branched urethanes in which the fraction of the aliphatic polyesters of the copolymers have the structure given above, and also blends of these

polyester resins with unmodified or modified polysaccharides, with water-vapour barrier properties of the type defined above, also fall within the scope of the invention.

Examples of bicarboxylic acids usable are succinic, adipic, pimelic, suberic, azelaic, sebacic, brassilic, undecandioic and dodecandioic acids, and dimeric acids; examples of hydroxy-acids which may be used are glycolic, hydroxybutyric, hydroxypropionic, hydroxycaproic, hydroxyvaleric, 7-hyroxyheptanoic, 8-hydroxyoctanoic, 9-hydroxynonoic, 10-hydroxydecanoic and 13-hydroxy-tridecancarboxylic acids.

Examples of diols which may be used are 1,2-ethandiol, 1,4-butandiol, 1,6-hexandiol, 1,7-heptandiol, 1,8-octandiol, 1,9-nonandiol, 1,10-decandiol, 1,12-dodecandiol, 1,4-cyclohexandimethylol and 1,4-cyclohexandiol.

Diacids and dialcohols which come from renewable sources and which can be produced from fatty acids such as oleic and ricinoleic acids are preferred.

When the diol has less than 7 carbon atoms, the acid has a number of carbon atoms such that the half-sum of the carbon atoms of the diol and of the acid is equal to or greater than 6, more preferably equal or higher than 7. The same criterion applies when the bicarboxylic acid has less than 7 carbon atoms.

The polycondensation is performed at temperatures of between 180° and 230°C in the presence of known catalysts based on transition and rare-earth metals such as tin, titanium, antimony, zinc, etc.

In the case of copolymers formed by or containing units or sequences of units X and Y, the preparation is performed in accordance with known methods by polycondensation of the diacid and the diol in the presence of the preselected lactone or lactide.

The mean numeral molecular weight obtainable by polycondensation may go up to values of the order of 100000 but it is preferably kept between 45000 and 70000.

Mean numeral molecular weights of less than 10000 do not permit the production of products having mechanical properties of practical interest.

The molecular weight can be increased by post-condensation reactions, operating either in the fused state or in the solid state, in the presence of polyfunctional compounds having groups reactive with the terminal -OH groups of the polyester, such as aliphatic or aromatic diisocyanates.

For post-condensation reactions (upgrades) in the solid state, the reaction is carried out by placing the solid resin in granular form in contact with the polyfunctional compound at ambient temperature or at a temperature slightly below the melting point of the resin for a period of time sufficient to bring about the desired increase in molecular weight.

The polyfunctional compound is used in the molten state, or dispersed homogeneously on the solid resin. Preferably, however, it is mixed with the resin in the fused state, for example, in an extruder, with periods of less than 5 minutes spent in the extruder to prevent undesired cross-linking reactions.

The intrinsic viscosity (measured in chloroform at 25°C) is increased even beyond 1 dl/g. Preferably, it is brought to values greater than 0.7 dl/g and most preferably between 0.8 and 2.5 dl/g. The viscosity of the resin in the fused state after upgrading is generally between 2000 and 30000 Pas measured at 180°C and with a "shear rate" of 100 sec^{-1}.

Diisocyanates are the preferred polyfunctional compounds acting as chain extenders; they are used in sufficient quantity to react with the terminal -OH groups of the resin. The quantity is between 0.2 and 1 equivalent of -NCO isocyanic groups per -OH group of the resin.

The quantity, expressed by weight, is generally between 0.01 and 3% of the resin, preferably between 0.1 and 2%.

The preferred diisocyanates are hexamethylene diisocyanate, diphenylmethane diisocyanate and isophorone diisocyanate.

Examples of other polyfunctional compounds which may be used are epoxides such as epoxy ethane, and the dianhydrides of tetracarboxylic aromatic acids such as pyromellitic anhydride.

The dianhydrides and the epoxides are also generally used in quantities of between 0.01 and 2% by weight of the resin.

The following examples are provided by way of non-limiting illustration of the invention.

Example 1

A polybutylene sebacate film having an intrinsic viscosity of 1.26 measured at 0.2 g/dl in chloroform at 25°C (produced by polycondensation of sebacic acid with 1,4-butandiol) was

used for the production of organic refuse sacks, bags for growing plants in greenhouses with metering of micro-nutrients, mulching films, bags for vegetables and tubers which do not sweat, or for other specific applications in which a low permeability to water vapour is required. The permeability to water vapour of this film was 250 gx30μm/m^2 per day at 38°C and 90% RH.

The film for the different applications has been produced using a Ghioldi machine for film-blowing of 40mm of diameter and L/D= 30, a temperature of 125C and 60 rpm. The head of 100mm was cooled with air at 10C.

The polymer was also found particularly suitable for the production of products which are to come into contact with liquid foods, such as thermoformed cups, drinking straws and plates for fast-food.

In case of thermoformed sheets the sheets have been produced with a mono screw extruder of 30 mm of diameter and L/D=30, using a flat head of 20cm of width. The extrusion temperature was of 13oC, the thickness was of 700 microns. The sheet has been thermoformed at 80C in a round cup. In case of drinking straws a MAI machine was used of 60mm of diameter and L/D=25. The productivity at 150C was comparable with the one of polyethylene.

Example 2

A polyhexamethylene sebacate film having an intrinsic viscosity of 0.7 dl/g (produced by polycondensation of sebacic acid with 1,6-hexandiol and subsequent upgrading with 1,6-hexamethylene diisocyanate at 60°C to give an intrinsic viscosity of 1.3 dl/g) was used for the production of organic refuse sacks, bags for growing plants in

greenhouses with metering of micro-nutrients, mulching films, bags for vegetables and tubers which do not sweat, or for other specific applications in which a low permeability to water vapour is required as in example 1.

The permeability to water vapour of this film was 180 gx30μm/m^2 per day at 38°C and 90% RH.

Example 3

Polyhexamethylene sebacate having an intrinsic viscosity of 1.3 dl/g was used for the production of single-layer and multi-layer films and sheets and for the production of containers for foods and drinks. An HAAKE RHEOCORD machine was used with a diameter of 19mm and L/D=25. The flat head had a width of 10cm. The molten film was calandered on cardboard in order to obtain an extrusion coated product for food containers.

Comparison Example 1

Polyhexamethylene adipate was used for the production of films the permeability of which was 700 gx30μm/m^2 per day at 38°C and 90% RH.

Example 4

The barrier properties of the following polymers were measured: polyethylenesebacate polynonandiol sebacate, polydecandiol sebacate, polyoctandiol azelate, polyoctandiol brassilate.

The barrier properties, expressed as permeability to vapour in gx30μm/m^2 per day (measured with a Lissy L80-4000 vapour

permeability tester at 38°C and 90% RH) were 300, 109, 100, 168, and 98, respectively.

The biodegradation behaviour according to the method described in "Journal of Environmental Polymer Degradation" vol. 4, N1, 1996, p55-63 for all the polymers fell inside the range of less than 10% of biodegradation in 14 days and more than 90% in 6 months.

CLAIMS

1. Use of compositions comprising, in quantities sufficient to ensure the required performance, polyester resins with mean numeral molecular weights greater than 10000 formed by recurring units X = $[O-(CH_2)_n-OCO-(CH_2)_m-CO]$ and/or Y = $[O-(CH_2)_k-CO]$, where the half-sum of n + m is equal to or greater than 6 and k is a number equal to or greater than 6, or by copolymers comprising units and/or sequences having the formula $x_i[O-(CH_2)_{mi}-OCO-(CH_2)_{mi}-CO]$; y_j $[O-(CH_2)_{kj}-CO]$ where: i,j = 1-5; n_i = 2-22; m_i = 0-20; k_j = 1-21;

$\sum_{i=1}^{5} x_i + \sum_{j=1}^{5} y_j = 1$ and x_i and y_j vary between 0 and 1 and are molar fractions of the various units such that

$$\sum_{i=1}^{5} x_i \cdot \left(\frac{n_i + m_i}{2}\right) + \sum_{j=1}^{5} y_j \cdot k_j \geq 6,$$

or by recurring units Z= $[O-(CH_2)_a-OCO-(CH_2)b-CO]$ where a=2-3, b=7-11,
present in sufficient quantity to ensure good barrier properties and biodegradability of the resins for the manufacture of articles having a permeability to water vapour of less than 350 gx30μm/m^2 per day at 38°C and 90% RH, said articles showing decomposition in composting conditions on 30μm film of less than 10% in 14 days and more than 90% in six months.

2. Use according to Claim 1, in which the polyester resins have a melting point of between 60 and 110°C.

3. Use according to Claim 1, in which the polyester resin

is produced by polycondensation of bicarboxylic aliphatic acids with from 2 to 22 carbon atoms and of diols with from 2 to 22 carbon atoms, selected in a manner such that the half-sum of the number of carbon atoms relating to the acid and to the diol is greater than 6, or by polycondensation of hydroxy-acids, or by ring-opening of corresponding lactones or lactides having from 7 to 22 carbon atoms.

4. Use according to Claim 1, in which the diacids and the dialcohols are obtained from renewable sources.

5. Use according to any one of the preceding claims, in which the polyester resin is selected from polyethylene sebacate, polybutandiol sebacate, polyhexandiol azelate, polyhexandiol sebacate, polynonandiol azelate, polynonandiol sebacate, polyoctandiol azelate, polyoctandiol brassilate, polydecandiol sebacate and polydecandiol brassilate.

6. Use according to any one of the preceding claims, in which the polyester resin has an intrinsic viscosity greater than 0.7 dl/g in chloroform at 25°C.

7. Use according to any one of the preceding claims, in which the polyester resin is subjected to an upgrading process.

8. Use according to any one of the preceding claims, in which the polyester resin is a component of a blend of unmodified or modified polysaccharides.

9. Use according to any one of the preceding claims, in

which the polyester resin contains mineral or vegetable fillers and/or additives selected from lubricants, plasticizers, colourings, flavourings, perfumes, flame-proofing agents, stabilizers with regard to hydrolysis and to thermal degradation, and antioxidants.

10. Use according to any one of the preceding claims, in which the mean numeral molecular weight of the polyester resin is between 45000 and 70000.

11. Use according to Claim 1 wherein said articles are selected from:

- coatings which are produced by extrusion-coating, with water-vapour barrier properties, and which are usable for the packaging of fresh milk and dairy products, of meat, and of foods having high water content,
- multi-layer laminates with layers of paper, plastics material or paper/plastics material, aluminium and metallized films,
- films as such and multi-layer films with other polymer materials,
- sacks for organic refuse and for grass cuttings with periods of use longer than 1 week,
- single-layer and multi-layer food packaging comprising containers for milk, yoghurt, cheeses, meat and beverages, in which the layer in contact with the food or beverage is formed by the aliphatic polyester,
- composites with gelatinized or destructured starch, and/or complexed starch or natural starch as a filler,
- mono-directional and bi-directional films,
- semi-expanded and expanded products produced by

physical and/or chemical means, by extrusion, injection, or agglomeration of pre-expanded particles,

- expanded sheet and expanded containers for foods, for drugs, and for fast food,
- fibres, fabrics and non-woven fabrics in the hygiene, sanitary and clothing fields,
- composites with mineral and vegetable fillers,
- thermoformed sheets for the food or fast-food packaging fields,
- bottles for the food, cosmetics and pharmaceutical fields,
- fishing nets,
- containers for fruit and vegetables,
- extruded sections usable in the fast-food field and irrigation pipes in the agricultural field.

12. Use of polyester resins as defined in Claim 1 in blends with other biodegradable polymers having a permeability to water vapour greater than 300 gx30μm/m^2 per day at 38°C and 90% RH.

13. Use of polyester resins as defined in Claim 1 in blends with polylactic acid.

14. Use of polyester resins as defined in Claim 1 in blends with other non-biodegradable polymers, the said polymers having a permeability to water vapour of less than 300 gx30μm/m^2 per day at 38°C and 90% RH.

Docket No.: C13929/I22944

DECLARATION AND POWER OF ATTORNEY FOR UTILITY PATENT APPLICATION - JOINT

As the below named inventors, we hereby declare that:

Our residences, post office addresses, and citizenships are as stated below next to our names.

We believe that we are the original, first, and joint inventors of the subject matter which is claimed and for which a patent is sought on the invention entitled "Use of polyester resins for the production of articles having good properties as barriers to water vapour", the specification of which is attached hereto.

We hereby state that we reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We acknowledge the duty to disclose information that is material to the examination of this application in accordance with 37 Code of Federal Regulations § 1.56.

We hereby claim foreign priority benefits under 35 United States Code §§ 119 and 365 of any foreign application(s) for the patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Applications: No. T098A000729 of August 28, 1998 and No. T098A000907 of October 26, 1998

We hereby appoint the following attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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We hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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